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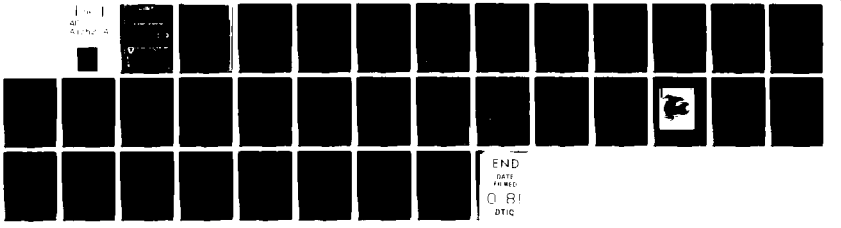

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CARS SPECTROSCOPY OF GUN PROPELLANT FLAMES

L. E. HARRIS
M. E. McILWAIN

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US ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND
LARGE CALIBER
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DOVER, NEW JERSEY

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Temperature measurements were made in a slightly fuel rich, premixed, propane/air reference flame and nitrate ester propellant flames burning in air at atmospheric pressure using Coherent Anti-Stokes Raman Scattering (CARS). Both single- and multiple-pulse CARS nitrogen spectra in the reference flame agreed with model spectra and the derived temperatures agreed with sodium line reversal values. Single pulse CARS nitrogen spectra obtained in the propellant flames were analyzed to give temperatures consistent with the values calculated using the NASA-Lewis thermochemical calculation. Comparison of a 0.1-second (cont)

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20. ABSTRACT (cont)

separated sequence of single pulse CARS spectra indicated turbulent air mixing in these propellant flames. The results demonstrated that temporal and spatially resolved temperature measurements could be determined in transient, turbulent flames.

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INTRODUCTION

Accurate in situ determination of gas and flame temperature and species concentrations are required for elucidating the complex dynamics involved in ballistic events. As new propellant materials are developed, these measurements can provide a means of evaluating their ballistic properties and are useful for comparison with ballistic computer calculations.

A wide data base of experimentally measured propellant flame temperatures and in situ species concentrations is nonexistent primarily because propellant flames are transient, turbulent, particle-laden, and luminous. These properties of propellant flames make conventional measurements, such as line reversal and Raman, difficult or impossible to perform. A relatively new technique [Coherent Anti-Stokes Raman Scattering (CARS)] may greatly facilitate in situ measurements. Recently CARS spectroscopy has been used to investigate not only stationary laboratory flames but also flames of practical interest (refs 1 through 4). The results of these studies indicate that CARS measurements can be performed upon nonideal flames such as those produced by propellants.

CARS PROCESS

The CARS process (fig. 1) involves the interaction of two high intensity laser beams (pump and Stokes beams) at angular frequencies ω_1 and ω_s. When the phase of the two frequencies is matched and the difference between ω_1 and ω_s is equal to a vibrational frequency of the gas being probed, an anti-Stokes photon is generated at

$$\omega_{as} = 2\,\omega_\ell - \omega_s \tag{1}$$

The intensity of the anti-Stokes emission is related to the induced polarization of the medium which is expressed as

$$P(\omega) = \chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 \tag{2}$$

where P is the induced polarization, χ is the dielectric susceptibility, and E is the electric field strength. It can be shown that for homogenous media the CARS process is proportional to the third order susceptibility and the power generated at the anti-Stokes frequency is

$$P\omega_{as} = A\left(\frac{\omega_{as}}{\eta_{as}}\right)^2 /3\chi^{(3)}/^2 P\omega^2_\ell\; P\omega_s \tag{3}$$

where A is a constant, η is the refractive index at ω_{as}, P is the laser power, and $/3\chi^{(3)}/$ is the magnitude of the total third order susceptibility. The total third order susceptibility of the medium is composed of frequency dependent (resonant) and independent (non-resonant) terms

$$/3\chi^{(3)}/^2 = \chi_R^{\,2} + 2\chi_R \bullet \chi_{NR} + \chi_{NR}^{\,2} \tag{4}$$

For a simple, damped harmonic oscillator χ_R is expressed as

$$\chi_R = K\Delta N_i \,/\, [2(\omega_{ij} - (\omega_\ell - \omega_s)) - i\Gamma_j] \tag{5}$$

where ω_{ij} is the ro-vibrational frequency, Γ_j is the Raman linewidth, ΔN_i is the population difference between the upper and lower states for a particular transition, and K is defined as

$$K = \frac{Nc^4}{K\omega_s^4}\left(\frac{d\sigma}{d\Omega}\right) \tag{6}$$

where N is the total number density, c is the speed of light, and $\frac{d\sigma}{d\Omega}$ is the Raman scattering cross section.

The above expressions assume that all frequencies are phase matched which can be experimentally accomplished in two ways for gaseous molecules. The ω_1 and ω_s beams can be combined on a straight line or at large angular separations called BOXCARS (ref 5). As shown in figure 2, the BOXCARS Phase matching conditions are satisfied by

$$2\,\eta_\ell\omega_\ell \cos\alpha = \eta_s\,\omega_s\cos\Theta + \eta_{as}\,\omega_{as}\cos\delta \tag{7}$$

where n_i is the refractive index at wavelength i.

Although the CARS process is related to conventional Raman scattering, the technique has the following advantages:

1. The light emitted during the CARS process is shifted to higher frequencies than ω_1, eliminating laser produced fluorescent interferences.

2. The process of mixing the various laser beams produces coherent (beam-like) emission which can be efficiently captured and detected.

3. The stimulated nature of the process greatly enhances the conversion efficiency of the blue shifted anti-Stokes emission so that it is several orders of magnitude greater in intensity than normal Raman.

4. The mixing geometry of the laser beams produces emission from discrete and easily identified volumes.

5. The pulse nature of the lasers used to generate the CARS process allows for sufficient temporal resolution to freeze out the chemical and fluid dynamics.

These aspects of CARS make the technique attractive for probing transient flames.

The primary purpose of these CARS experiments was to determine the reliability of the CARS technique for making temperature measurements and to demonstrate the feasibility of using the technique to probe transient, turbulent flames.

EXPERIMENTAL

CARS spectra are generated by use of the apparatus shown in figure 3. The pump laser beam is produced by a Quanta-Ray DCR-1A Nd/YAG laser. The output of the Nd/YAG laser at 1.06 microns (800 mj) is doubled to generate the pump beam at 5320 Å (250 mj) which has a bandwidth less than 2 cm^{-1}. The pump beam is separated from the primary beam using prisms. Forty percent of the pump beam is split off (BS_1) to pump a dye laser to generate the Stokes beam. The dye laser is operated broadband at 6073 Å (10 mj) with a 150 cm^{-1} bandwidth. In BOXCARS configuration the pump beam is split into two components which are spatially separated to form an angle, 2α, when focused. The Stokes beam is introduced at a second angle, θ, in the plane of the pump beams so that phase matching occurs. CARS is generated in the region where the three beams cross at a third angle, γ. To achieve BOXCARS geometry, the pump beam is split using a 50 % beamsplitter (BS_2). The ω_1 beam is reflected from a dichroic (DC) which transmits the Stokes beam, and ω_1' is separately reflected along another path so that the pump beams are separated at the focusing lens. The BOXCARS signal is generated along ω_1' and, after dispersal and spatial separation with an aperture, is focused on the slits of a monochrometer fitted with a PAR SIT detector. The signal from the detector is subsequently sent to an OMA2 for processing.

In these experiments a 250 mm focusing lens was used with a pump beam crossing angle of 5°. This configuration has been shown experimentally (ref 5) to give a spectral resolution near 1 mm^3. A 1/4 meter monochromator equipped with an 1800 line per/mm grating gave an experimental spectral resolution of 8.00 cm^{-1} with 2.5 cm^{-1} per channel.

Measurements were made on a premixed propane/air flame maintained on a 1.6-cm-diameter circular burner surface constructed of a matrix of steel syringe needles 0.2 cm outer diameter. The flame was stabilized by flowing N_2 through a 2-cm-diameter concentric outertube. The flow rates of propane and air were adjusted to approximately 130 and 2000 cm^3 per min, respectively.

The propellant used was U.S. Navy SGP-38, which has the composition given in table 1. The propellant samples, in the form of right-circular cylinders 12-mm long, were burned in air.

Shadowgraphs of the propellant flame were produced with an argon spark of approximately 1.0 microsecond duration with a Z-fold optical configuration.

Temperature Determination

In most air fed flames, nitrogen is a major inert constituent in the burning gases issuing from the burner. An abundance of nitrogen is also predicted in propellant flames by thermochemical calculations (table 2). Nitrogen is a convenient molecule to detect and use as an in situ gas thermometer. The actual process of measuring a nitrogen CARS spectrum does not directly yield the gas temperature. The shape of the spectral profile is indicative of the gas temperature. The change of the spectral profile must be calculated. A computer model

is used to produce synthetic spectra. The input to this program is the gas temperature,the molecular constants of the probe gas, and the value for the nonresonant susceptibility which accounts for the influence of other flame constituents observed in experimental spectra. The scheme used to generate these computer synthesized spectra is described in reference 6. The energy of the vibrational and rotational transitions are calculated and individual level populations are assumed to follow a Boltzman distribution.

These energies and populations are convoluted by equation 3 and the resulting intensities are plotted as a function of $\omega_1-\omega_s$. These synthetic spectra can be compared directly to experimental spectra as displayed in figure 4 or in cases where the hot band [(2-1) band] is present, its height is compared to the curve shown in figure 5 to estimate the gas temperature.

Table 1. SGP-38 propellant composition

Component	Weight %
Nitrocellulose (12%)	47.5
Metriol trinitrate	39.4
Triethyleneglycol dinitrate	10.0
Ethyl centralite	1.0
Lead beta resorcilate	1.0
Cryolite	1.0
Cordella wax	1.0

Table 2. Thermochemical calculations

P = 1 ATM T = 2600 K

	CO	CO_2	H_2	H_2O	N_2
Mole fraction	0.43	0.07	0.18	0.19	0.10

Flame Measurements

To assess the ability of our computer model to accurately determine flame temperatures, a number of spectral measurements were determined at various positions in a slightly fuel rich propane/air flame. CARS spectra were collected both during a single laser shot and as an average of single shot spectra. Single shot spectra (fig. 4) were obtained, while averaged spectra resembled that displayed in figure 6.

The modeled spectrum (figs. 4 and 6) is in agreement with the average spectra and, considering the low signal to noise for the single shot spectrum, the fit is acceptable.

The gas temperature is displayed in figure 7 as a function of position in the premixed flame. As expected for premixed flames maintained on a flat flame burner, the temperature remains nearly constant across the width and height of the flame. Measured temperatures can be compared to that calculated for a stoichiometric flame (2250 K) and determined for a propane/air diffusion flame (2150 K).

To determine the accuracy of the CARS temperature measurements additional measurements were made using the sodium line reversal technique. The CARS and sodium line reversal temperatures are compared in figure 8. The average difference between temperatures determined by the two methods at the same point in the flame is 30.1 K.

To determine the feasibility for CARS temperature determinations in non-ideal flames, CARS spectra were recorded in propellant flames at various positions along the flame centerline above the burning propellant surface. For simplicity the experiments were carried out in air at atmospheric pressure. The single pulse spectra (fig. 9) were produced at 0.1 second intervals approximately 63 mm above the propellant surface. The dramatic difference in spectral profile and analyzed temperatures were an indication of turbulence and mixing of the hot gases with air. A CARS spectrum was also produced at approximately 10 mm above the surface in a single pulse (fig. 10). Although the spectrum was reduced in intensity and had a much lower signal to noise ratio, it was analyzed to yield a gas temperature.

DISCUSSION

The reference flame results confirm the usefulness of CARS spectroscopy for flame temperature determination. The 2050 K maximum temperature measured for the premixed propane/air flame is not unrealistic considering the position above the burner where measurements were made, the stoichiometry of the gases, and the burner design. The average agreement with Na line reversal temperatures is $\pm$ 30 K. This is consistent with other CARS measurements recorded at lower temperatures and those reported in the literature (ref 4), which determined the error for temperatures evaluated in this manner to be $\pm$ 50 K. Using this error the propane/air flame temperatures are consistent with those reported for a propane diffusion flame (ref 4). The similarity between diffusion and premixed flame temperatures is not surprising. It is expected that only the positions of maximum temperature are dependent upon flame type, since the same chemistry must occur in both flame types (ref 7). The agreement with reported flame temperatures, calculated values, and those determined in this work increases the validity of CARS temperature determination.

The initial attempt to use CARS to investigate actual propellant flames was successful. The nitrogen CARS signal intensity was sufficient to obtain flame temperature even for the spectrum recorded near the propellant surface. At this

position one would expect that all nitrogen present was produced by the propellant combustion, since the mass flow from the surface would be sufficient to retard air from diffusing into this region. Typically, the materials produced considerable amounts of soot and incandenscence on burning. Yet, the measurements were relatively free from interferences, and the only interference observed was due to large particles blocking one or more beams.

The high temperature measured near the propellant surface, 2500 K, is in satisfactory agreement with the calculated flame temperature of 2600 K. The range of temperature determined from spectra obtained at 63 mm above the propellant surface is indicative of turbulent flow and mixing of surrounding air with hot combustion gases. The mixing process is further substantiated by shadowgraphs such as that shown in figure 11, which display eddies and dark zones at this position in the propellant flame.

The unconfined burning of the propellant in air limits the interpretation of the measured flame temperature in relation to combustion models such as the Parr-Crawford nitrocellulose model, (ref 8). The influence of air on these reaction zones alters the chemical reactions which occur and affect heat feedback to the solid surface. Further work is required to better understand the influence of air in this reaction scheme.

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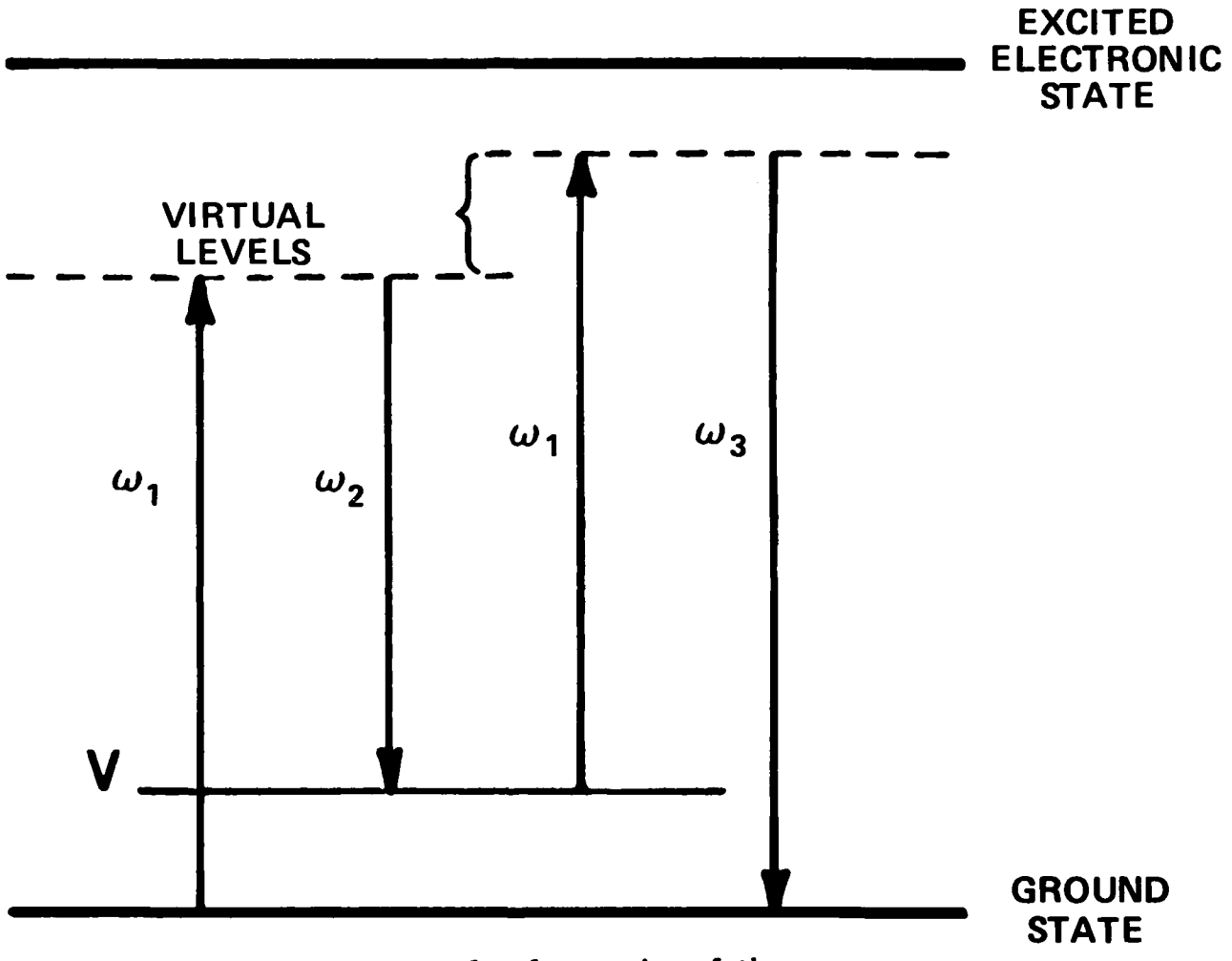
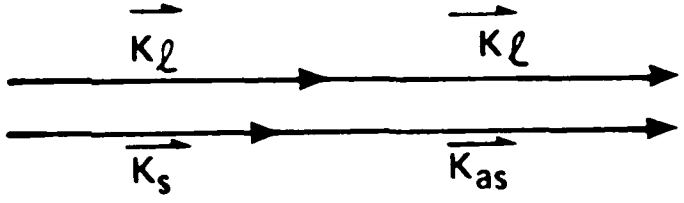


ω_l, ω_s and ω_{as} = angular frequencies of the pump, strokes and CARS beams, respectively

Figure 1. CARS energy level diagram

COLLINEAR



BOXCARS

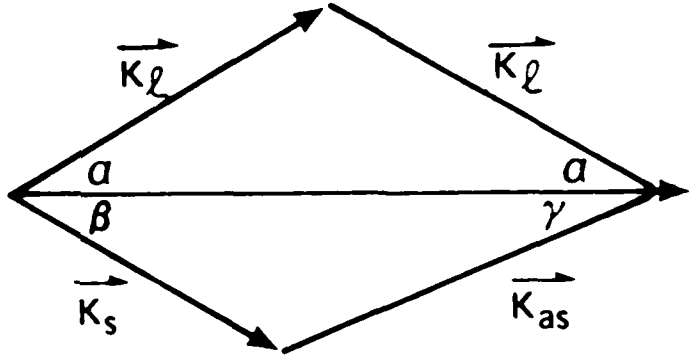


Figure 2. Phase matching configuration for gases

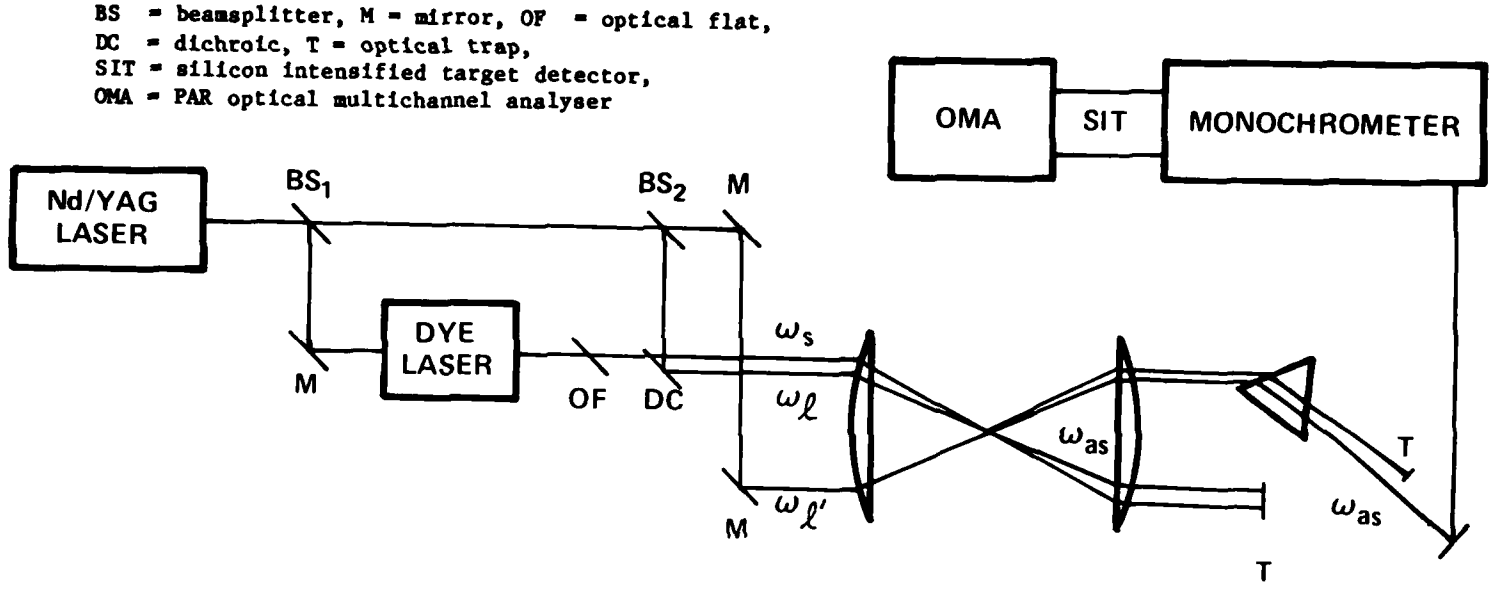


Figure 3. CARS instrumental schematic

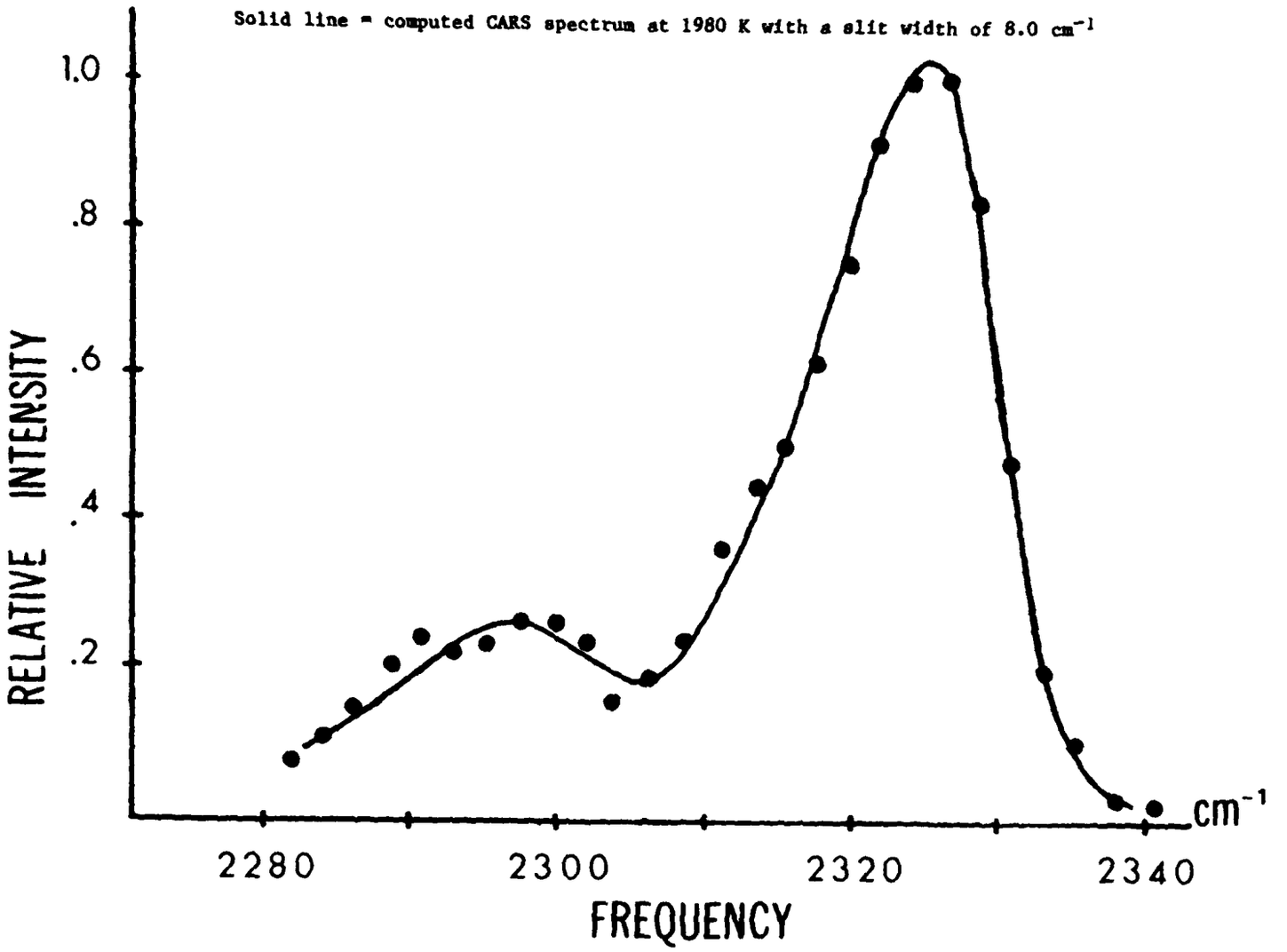


Figure 4. CARS nitrogen spectrum from a premixed propane and air flame taken using a single pulse I_{max} - 80 counts

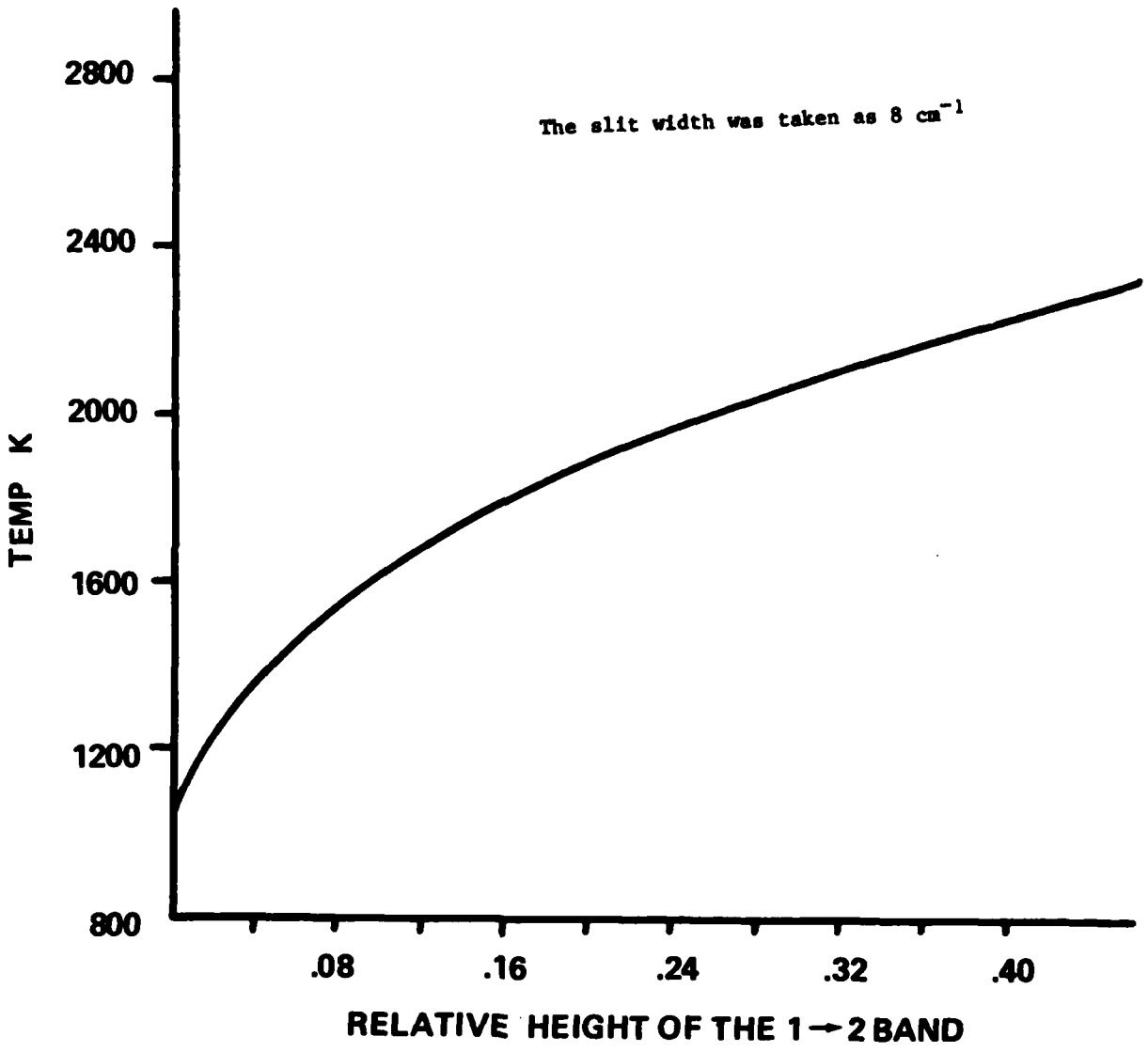


Figure 5. Height of the 1-2 band (relative to a 0-1 band normalized to one) with respect to change in temperature

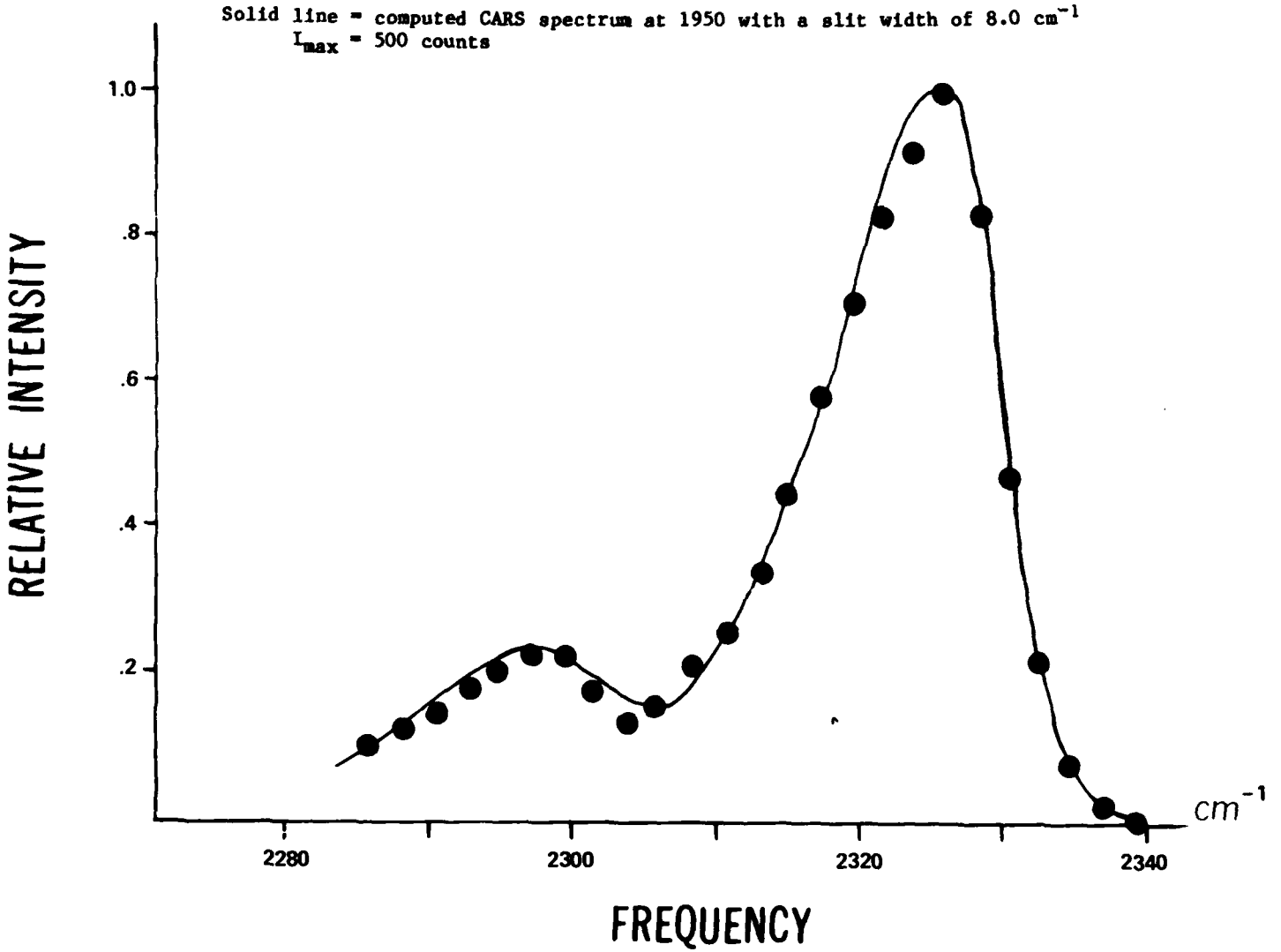
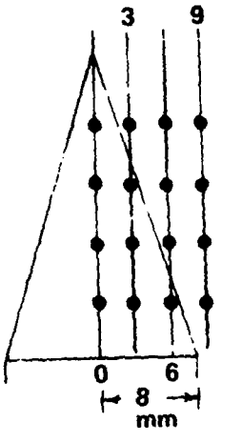


Figure 6. CARS nitrogen spectrum of premixed propane and air flame obtained by averaging ten pulses

Values in parenthesis represent horizontal displacement (mm) from the burner centerline. Burner halfwidth is 8 mm.



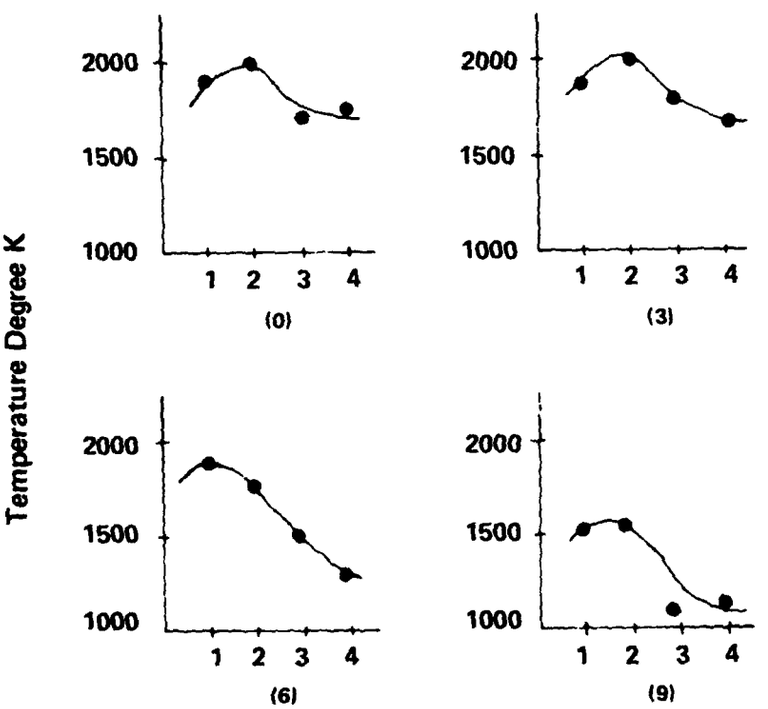


Figure 7. Variation of temperature with respect to vertical position above the burner surface (mm) in a premixed propane and air flame

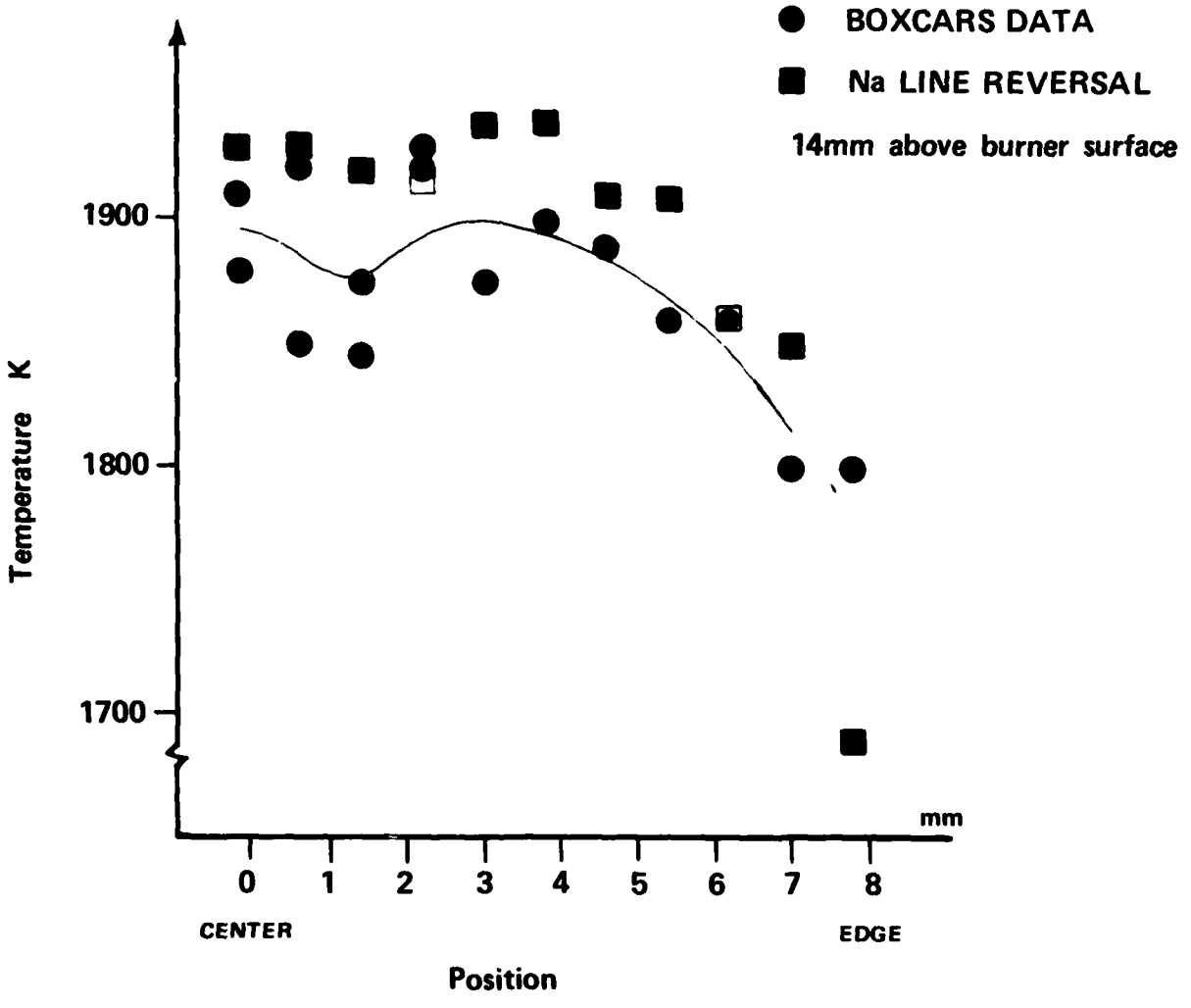


Figure 8. Temperature determined by CARS and Na line reversal in a premixed propane and air flame with respect to horizontal displacement from the burner centerline (mm)

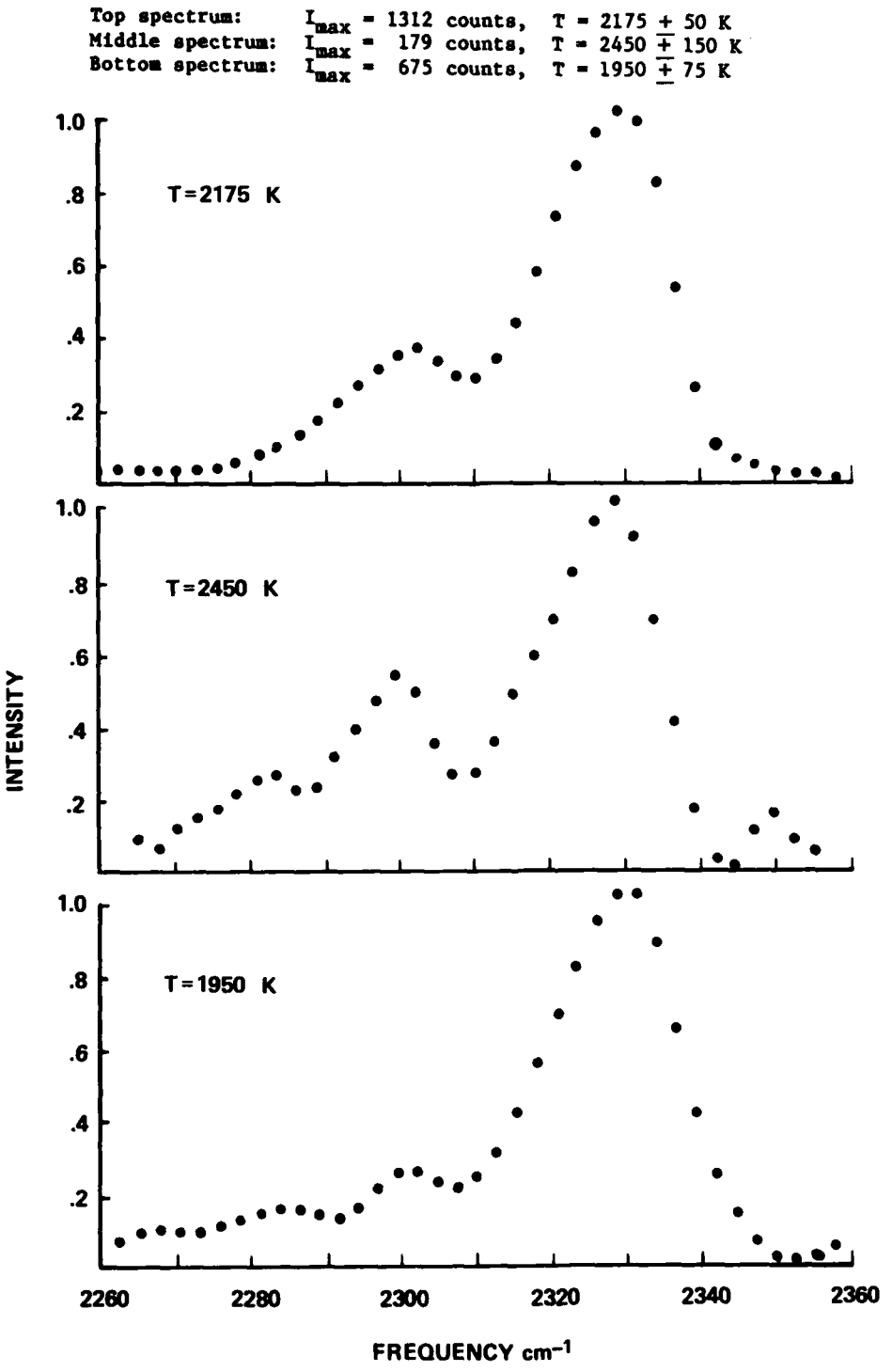
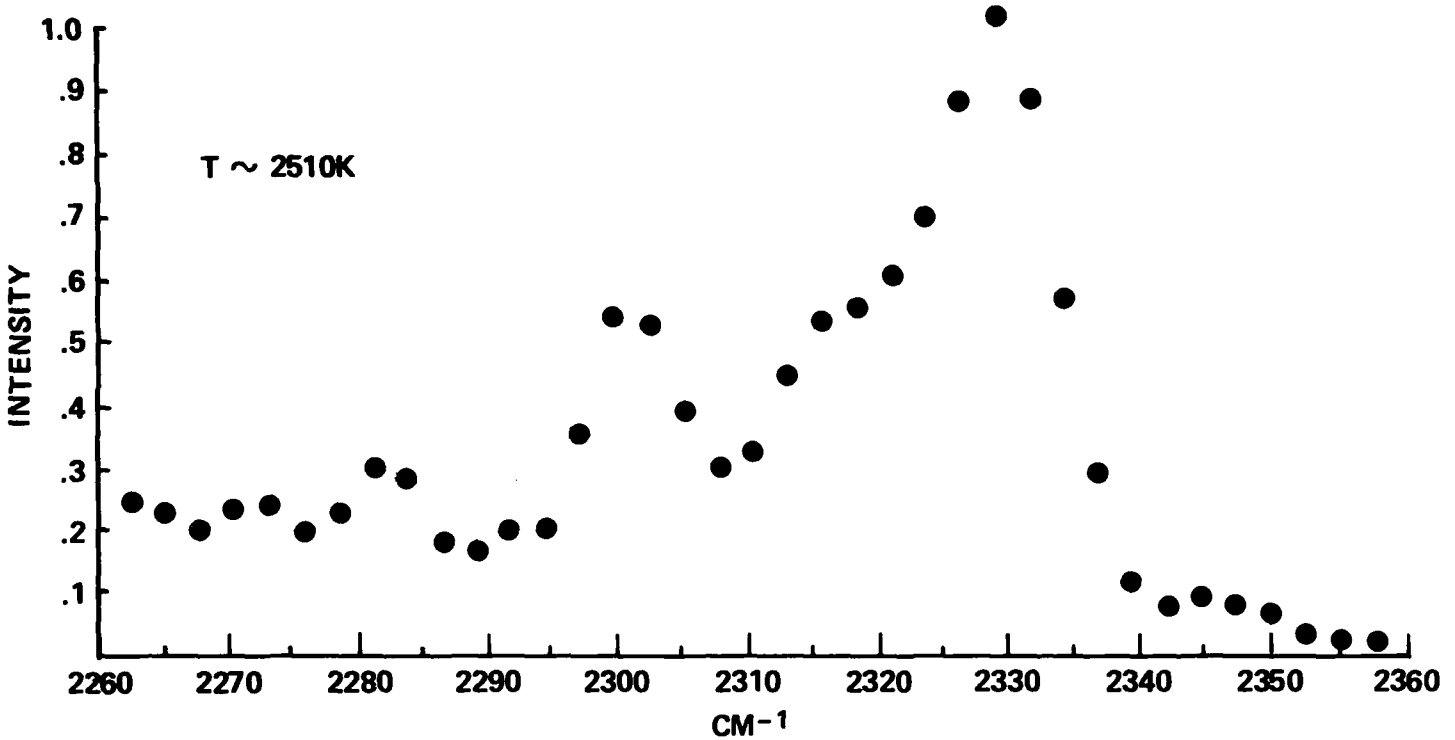


Figure 9. CARS nitrogen spectra of SGP-38 propellant flame burning in air taken using single pulses 0.1 sec apart 63 mm above the centerline of the propellant surface



I_{max} = 212 counts, T = 2500 $\pm$ 150 K

Figure 10. CARS nitrogen spectrum of SGP-38 propellant flame burning in air taken using single pulse 10 mm above the centerline of the propellant surface

Figure 11. Shadowgraph of propellant flame taken using an Argon spark of one microsecond duration

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ATTN: Code 5B331/R. S. Lazar
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Commander
Naval Weapons Center
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C. Thelen
China Lake, CA 93555

Commander
Naval Research Laboratory
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Washington, DC 20375

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Naval Postgraduate School
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B. T. Wolfson
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L. Caveny
Bolling AFB, DC 20332

AFRPL (DYSC)
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J. N. Levine
Edwards AFB, CA 93523

National Bureau of Standards
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Propulsion Division
23146 Cumorah Crest
Woodland Hills, CA 91364

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N. Hollywood, CA 91602

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Elkton Division
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Elkton, MD 21921

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Huntsville Division
ATTN: D. Flanigan
R. Glick
Technical Library
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Thiokol Corporation
Wasatch Division
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Technical Library
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Brigham City, UT 84302

TRW Systems Group
ATTN: H. Korman
One Space Park
Redondo Beach, CA 90278

United Technologies
Chemical Systems Division
ATTN: R. Brown
Technical Library
P.O. Box 358
Sunnyvale, CA 94086

Universal Propulsion Co
ATTN: H. J. McSpadden
1800 W. Deer Valley Road
Phoenix, AZ 85027

Battelle Memorial Institute
ATTN: Technical Library
R. Bartlett
505 King Avenue
Columbus, OH 43201

Brigham Young University
Dept of Chemical Engineering
ATTN: M. W. Beckstead
Provo, UT 84601

California Institute of Tech
204 Karmar Lab
Mail Stop 301-46
ATTN: F. E. C. Culick
1201 E. California Street
Pasadena, CA 91125

Case Western Reserve Univ
Division of Aerospace Sciences
ATTN: J. Tien
Cleveland, OH 44135

Georgia Institute of Tech
School of Aerospace Eng
ATTN: B. T. Zinn
E. Price
W. C. Strahle
Atlanta, GA 30332

Institute of Gas Technology
ATTN: D. Gidaspow
3424 S. State Street
Chicago, IL 60616

Johns Hopkins University/APL
Chemical Propulsion Info Ag
ATTN: T. Christian
Johns Hopkins Road
Laurel, MD 20810

Massachusetts Inst of Tech
Dept of Mech Engineering
ATTN: T. Toong
Cambridge, MA 02139

Pennsylvania State University
Applied Research Lab
ATTN: G. M. Faeth
P.O. Box 30
State College, PA 16801

Pennsylvania State University
Dept of Mechanical Engineering
ATTN: K. Kuo
University Park, PA 16801

Pennsylvania State University
Dept of Material Sciences
ATTN: H. Palmer
University Park, PA 16801

Princeton Combustion Research
Laboratories
ATTN: M. Summerfield
1041 U.S. Highway One North
Princeton, NJ 08540

Princeton University
Forrestal Campus
ATTN: I. Glassman
Technical Library
P.O. Box 710
Princeton, NJ 08540

Purdue University
School of Mechanical Eng
ATTN: J. Osborn
S. N. B. Murthy
TSPC Chaffee Hall
W. Lafayette, IN 47906

Rutgers State University
Dept of Mechanical and
Aerospace Engineering
ATTN: S. Temkin
University Heights Campus
New Brunswick, NJ 08903

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J. Barker
D. Golden
333 Ravenswood Avenue
Menlo Park, CA 94025

Stevens Institute of Tech
Davidson Library
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Hoboken, NJ 07030

United Technology
ATTN: Alan Ecbreth
Research Center
East Hartford, CT 06108

Director
U.S. Army Materiel Systems
Analysis Activity
ATTN: DRXSY-MP
Aberdeen Proving Ground, MD 21005

Commander/Director
Chemical Systems Laboratory
U.S. Army Armament Research and
Development Command
ATTN: DRDAR-CLJ-L
DRDAR-CLB-PA
APG, Edgewood Area, MD 21010

Director
Ballistics Research Laboratory
U.S. Army Armament Research and
Development Command
ATTN: DRDAR-TSB-S
DRDAR-BLP, L. Watermier
A. Barrows
C. Nelson
J. Vanderhoff
J. Anderson
Aberdeen Proving Ground, MD 21005